

WHAT IS CLAIMED IS:

1 1. A process for the preparation of organic isocyanates, said
2 process comprising:

- 3 a) mixing an organic formamide compound or its amine and
4 formate precursors with a diorganocarbonate to form a
5 reaction mixture;
6 b) subjecting said reaction mixture to an elevated temperature
7 sufficient to generate the isocyanate corresponding to said
8 organic formamide compound; and
9 c) isolating said isocyanate from said reaction mixture.

1 2. The process of claim 1, wherein said organic formamide
2 compound is one of the formula



4 where n is an integer from 1 to 10 and R is an organic radical.

1 3. The process of claim 2, wherein R comprises an optionally
2 substituted C_{1-20} alkyl, C_{2-20} alkenyl, C_{2-20} alkynyl, C_{4-20} cycloalkyl, C_{4-20}
3 cycloalkenyl, C_{6-30} aryl, C_{7-30} aralkyl, C_{7-30} alkaryl group, a silane or siloxane or
4 oligomer thereof wherein formamide group(s) are bound to an Si-bound hydrocarbon
5 linking group, and wherein each of the above R may contain one or more chain or
6 ring heteroatoms.

1 4. The process of claim 2, wherein R is selected from the group
2 consisting of optionally substituted phenyl, diphenylmethane, and tolyl groups.

1 5. The process of claim 4, wherein R is the 2,4-bis(N-formamide)
2 of toluene diamine.

1 6. The process of claim 1, wherein steps a) and b) are combined
2 such that the reaction temperature of step a) is maintained at a temperature wherein
3 isocyanate is produced directly.

1 7. The process of claim 6, wherein the temperature of steps a)
2 and b) are in the range of 150°C to 240°C.

1 8. The process of claim 1, wherein the temperature of step a) is
2 maintained below 190°C and at a first temperature such that no substantial
3 production of isocyanate occurs, said process further comprising:

4 a)i) isolating an intermediate, isocyanate precursor mixture
5 containing a carbamate group-containing reaction
6 product, and thermolyzing said isocyanate precursor
7 mixture at a second, higher temperature to obtain the
8 isocyanate corresponding to said organic formamide.

1 9. The process of claim 8, wherein said isocyanate precursor
2 reaction mixture comprises in excess of 80 mol percent of the carbamate
3 corresponding to said organic formamide, said mol percent based on the total of
4 mols of isocyanate, carbamates and isocyanate/carbamate contained in said
5 isocyanate precursor reaction mixture.

1 10. The process of claim 1, wherein said diorganocarbonate is
2 selected from aliphatic, cycloaliphatic, aryl, and mixed aliphatic/aryl or
3 cycloaliphatic/aryl carbonates.

1 11. The process of claim 1, wherein said diorganocarbonate is
2 diphenylcarbonate.

1 12. The process of claim 2, wherein R is aliphatic or
2 cycloaliphatic, said process further comprising adding to said reaction mixture an
3 effective carbamide-cleaving amount of a metal catalyst.

1 13. The process of claim 1, wherein the ratio of mols of
2 diorganocarbonate to organic formamide is greater than 1:1 based on mols of
3 formamide groups.

1 14. The process of claim 1, wherein the ratio of mols of
2 diorganocarbonate to mols of formamide groups is 2:1 to 5:1.

1 15. The process of claim 1, wherein one or more organic di- or
2 polyamines, an organoformate ester, and diorganophenol carbonate comprise said
3 reaction mixture.

1 16. The process of claim 15, wherein said organo group of said
2 organoformate is the same as at least one of the organo groups of said
3 diorganocarbonate, said organo groups selected from the group consisting of C₁₋₂₀
4 aliphatic, C₄₋₂₀ cycloaliphatic, C₆₋₂₀ aryl, C₇₋₃₀ aralkyl, and C₇₋₃₀ alkaryl groups, their
5 heteroatom substituted analogs, and mixtures thereof.

1 17. The process of claim 15, wherein said organo groups are
2 selected from the group consisting of C₁₋₂₀ alkyl, C₅₋₈ cycloalkyl, and optionally
3 substituted C₆₋₁₀ aryl.

1 18. The process of claim 15, wherein all organo groups are
2 phenyl.

1 19. The process of claim 15, which is a continuous process
2 wherein organoformate is removed from said reaction mixture and recycled to said
3 reaction mixture with additional organic di- or polyamine.

1 20. A process for the direct manufacture of an organic isocyanate
2 from the corresponding formamide, said process comprising:

3 a) reacting an organic formamide containing from 1 to 10
4 formamide groups per molecule with from 1 to about 10 mol
5 of diorganocarbonate per mol of formamide groups to form a

6 reaction mixture, said reacting taking place at a temperature
7 such that themolysis of products contained in said reaction
8 mixture generates the isocyanate corresponding to said
9 organic formamide;
10 b) separating said isocyanate from said reaction mixture.

1 21. The process of claim 20, wherein said isocyanate separated
2 from said reaction mixture also contains partially thermolyzed products containing
3 carbamates corresponding to said organic formamide and/or mixed
4 isocyanate/carbamide compounds corresponding to said organic formamide, said
5 process further comprising:

- 6 b)i) further thermolyzing said partially thermolyzed
7 products to form additional isocyanate corresponding
8 to said organic formamide; or
9 b)ii) returning said partially thermolyzed products to said
10 reaction mixture; or
11 b)iii) performing both of b)i) and b)ii).

1 22. The process of claim 20, wherein diphenylcarbonate is
2 employed as said diorganocarbonate, reaction takes place in phenol solvent, and said
3 isocyanate separated from said reaction mixture contains phenol and phenol formate
4 ester, said process further comprising recycling said phenol formate ester by reacting
5 said phenol formate ester with an organic amine corresponding to said organic
6 formamide to form said organic formamide.

1 23. A continuous process for producing organic isocyanates, said
2 process comprising:
3 a) reacting an aryl di- or polyformamide or an amine and
4 formate precursor thereof, with diphenyl carbonate at a
5 temperature at least sufficient to form a reaction mixture
6 containing O-phenylcarbamates corresponding to said aryl di-
7 or polyformamide;

- 8 b) thermolyzing all or a portion of said reaction mixture to
9 generate an isocyanate-containing mixture comprising organic
10 isocyanate(s) corresponding to said aryl di- or polyformamide;
11 c) separating said organic isocyanate from said isocyanate-
12 containing mixture to obtain a purified organic isocyanate,
13 and an organic isocyanate depleted mixture c)i);
14 d) separating phenol from said isocyanate-containing mixture or
15 from said organic isocyanate-depleted mixture to form a
16 phenol-depleted mixture d)i);
17 e) separating carbamates and carbamate/isocyanates from said
18 isocyanate-containing mixture or from said mixture c)i), or
19 d)i) and further processing said carbamates and
20 carbamate/isocyanates by one or both of
21 e)i) further thermolyzing to form said organic
22 isocyanate(s) corresponding to said aryl di- or
23 polyformamide; or
24 e)ii) cycling said carbamates and/or said
25 carbamate/isocyanates into said reaction mixture of
26 step a),
27 to form a carbamate-depleted mixture e)iii
28 f) separating from one or more of said isocyanate-containing
29 mixture, c)i, d)i), or e)iii) phenol formate ester, and
30 g) optionally reacting said phenol formate ester with an organic
31 amine to form the formamide corresponding to said organic
32 amine.

1 24. The process of claim 23, wherein said step of thermolyzing
2 takes place at a temperature of from 150°C to 240°C.

1 25. A process for the preparation of an O-organocarbamate, said
2 process comprising reacting an organic formamide or its amine and formate
3 precursors with a diorganocarbonate at a temperature below that at which significant

- 4 thermolysis of O-organocarbamate to isocyanate occurs, and separating said O-
- 5 organocarbamate from other reaction products.

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